

Process for manufacturing (U,Pu)O₂ mixed oxide nuclear
fuel pellets from non-free-flowing UO₂ powder

The present invention relates to a process for
5 manufacturing a (U,Pu)O₂ mixed powder from non-free-
flowing UO₂ powders.

The manufacture of fuel for light-water
reactors, based on uranium and plutonium oxides,
generally called MOX fuel, has been the subject of
10 various developments associated with the desire to to
recycl plutonium recovered during spent fuel
reprocessing.

The manufacture and irradiation of MOX fuel in
light-water reactors are now considered to be a
15 solution for providing reasonable resistance to the
proliferation of plutonium present in a form separated
from the fission products, whether this plutonium is
either of civilian or military origin.

Several processes for manufacturing MOX fuel
20 have been developed over the last two decades, some of
the processes involving the complete milling of the UO₂
and PuO₂ powders in order to provide an intimate blend,
while others are limited to milling only a fraction of
these powders.

25 The MIMAS (standing for MICronization and
MASTER blend) process, which was developed by the
Applicant of the present invention (see figure 1),
comprises the micronization, by milling, of only a
30 fraction of the final blend and uses two successive
blending operations to achieve isotopic homogenization
and to take advantage of the use of free-flowing UO₂
incoming products (especially to ensure that the dies
of the presses used for pelletizing are properly
filled). Using free-flowing UO₂ powders in the second
35 blending operation and limiting the milling to only the
first blending operation simplify the manufacture (for
example by dispensing with the operations of
precompacting/granulating or spheroidization of the

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mixed oxide blend) and have greatly simplified, at the start of industrial implementation, the qualification of MOX fuel by users and the licensing process by the Nuclear Safety Authorities (thanks to the similarity in behavior between this MOX fuel and UO₂ fuel).

Various versions of the MIMAS process have been applied, sometimes under names different from MIMAS, but all characterized by two successive blending operations, the second of which uses free-flowing UO₂.

UO₂ which serves as feed material in the manufacture of enriched-uranium fuel and, in the great majority of cases, in the manufacture of MOX fuel, is obtained by the conversion of uranium hexafluoride. There are industrial conversion processes which produce free-flowing UO₂ powder. This is especially the case with two industrial conversion processes using a wet route, known in the art by the respective names "AUC", coming from the intermediate product (Ammonium Uranyl Carbonate), and "TU2", coming from the uranium transformation unit in which the conversion is carried out. One of the drawbacks of these wet conversion processes is the production of a large amount of liquid effluents which have to be treated before discharge. The wet conversion processes, some of which do not produce free-flowing UO₂, are gradually being replaced with dry processes which allow the gaseous effluents to be recycled but which generally produce non-free-flowing UO₂ powder.

For the purpose of diversifying the sources of UO₂ powder for manufacturing MOX fuel by MIMAS-type processes, it is therefore useful to be able to employ non-free-flowing UO₂ powders.

Non-free-flowing UO₂ powder conditioning processes, for transforming it into free-flowing UO₂ granules, and therefore having properties suitable for feeding a pelletizing press, are known. Various mechanical granulation processes, such as precompaction-granulation or agglomeration-

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spheroidization, have been developed and are used on an industrial scale in UO_2 fuel manufacturing plants.

Experience has shown that these granulation processes produce granules of insufficient mechanical strength for correct implementation of the second blending operation which characterizes the MIMAS processes and similar processes. Under the optimum operation of the second blender, the granules are damaged and the flowability of the secondary blend is impaired: the fuel pellets which result therefrom suffer from major defects (excessive variability in the physical properties of the product, local differential shrinkage defects, etc.). Alternatively, if the method of operating the second blender is modified so as to achieve gentle mixing of the powders to be blended, or if the apparatus used for the second blending is modified for the same purpose, the uniformity of distribution of the plutonium within the fuel may be impaired and the MOX pellets thus produced no longer meet the maximum plutonium content variability criteria.

To avoid the abovementioned drawbacks, the process for manufacturing MOX fuel from non-free-flowing UO₂ powder, which is the subject matter of the invention, comprises a mechanical granulation treatment of the non-free-flowing UO₂ powder, which does not modify the chemical properties (such as a stoichiometry) and morphological properties (such as the particle size) of the UO₂ powder, but which does nevertheless ensure the mechanical strength and flowability that are required to successfully carry out the second blending operation and the pelletizing operation, respectively.

The invention thus obviates the need to supply
35 the MIMAS-type processes with free-flowing UO₂ powders
as feed materials.

According to one advantageous method of implementing the invention, non-free-flowing UO₂ powder is used, one part of which is used, as it is, for

incorporation in the first blend and one part of which undergoes a granulation treatment before being incorporated into the second blend.

In a variant, as a nonlimiting example, said 5 granulation treatment may also be applied to the non-free-flowing UO_2 fraction fed in the first blend

In order to avoid the drawback of the abovementioned lack of mechanical strength of UO_2 granulated by one of the usual conditioning processes, 10 the mechanical treatment according to the invention is carried out either by forcing the non-free-flowing UO_2 powder through a screen or sieve, or by compressing this powder into tablets under a high pressure, as required for obtaining suitable non-friability 15 properties, and then crushing said tablets. When necessary, one or more binders and/or lubricants may be added beforehand to the UO_2 powder.

Further details and features of the invention will become apparent from the claims and from the 20 description of the drawings, which are appended to the present specification and which illustrate, by way of nonlimiting examples, the manufacturing process according to the invention.

Figure 1 shows schematically the steps in the 25 manufacture of mixed oxide fuel according to a known process of the MIMAS type.

Figure 2 shows schematically the steps in the manufacture of mixed oxide fuel according to a process of the invention.

30 Figure 3 shows schematically variants of the process according to the invention.

In the various figures, the same reference notations denote identical or similar components.

The process of the invention, for the use of 35 non-free-flowing UO_2 powder, comprises basically a process for the manufacture of $(U, Pu)O_2$ mixed oxide fuel pellets, that is to say overall (figure 2):

- dosing and first blending (step 1) of PuO₂ powders and/or UO₂ powders and/or fuel manufacturing scrap;
- micronization (step 2) of this first blend, particularly by milling, and forced sieving (step 3) of its product, for example through a 250 µm screen mesh;
- additional dosing and second blending (step 4) of the first blend thus treated, UO₂ and, where appropriate, fuel manufacturing scrap;
- addition, and blending with the resulting second blend of one or more lubricants and/or poreformers (step 5), the latter step possibly being completely or partly combined with step 4;
- compression (step 6) of the second blend into pellets using pelletizing presses ; and
- sintering (step 7) of the pellets thus formed, preferably in an atmosphere of moistened argon (or nitrogen) and hydrogen.

This mixed oxide fuel pellet manufacturing process may also usually include, for the pellets thus obtained, steps of:

- dry grinding (step 8);
- visual inspection (step 9);
- stacking up to length (step 10);
- loading the pellets into a cladding and welding the latter so as to form a fuel rod (step 11, figure 1);
- pressurizing the rods;
- nondestructive testing/examination of the rods (step 12); and
- assembling of the rods (step 13).

Said process of the invention furthermore includes (figure 2) a prior mechanical granulation treatment of all or part of the nonflowing UO₂ (step 29). This treatment may comprise, for example:

- either (figure 3) steps of compressing the non-free-flowing UO₂ into tablets (step 30) and of

crushing these tablets (step 31) and, where appropriate, of sieving the crushed material (step 32) in order to form free-flowing granules having properties suitable for being incorporated as the basic constituent in the second blending operation (step 4) or, in a variant, in both blending operations (steps 1 and 4), while maintaining the original chemical composition and original particle size of the original UO_2 ;

- or an agglomeration/precompaction/granulation step by forcing the non-free-flowing UO_2 powder through a screen or sieve (step 29), the amount of additive(s), the mesh size of the screen or sieve and the pressure exerted on the powder being adjusted in order to form granules having the suitable properties described above.

A few nonlimiting parameters of the pellet manufacturing process are given below by way of example:

- batch/campaign operation rather than continuous operation;
- plutonium content of the first blend: 20 to 40% (step 1);
- milling (step 4) in 60 kg batches for a minimum effective time of 5 hours;
- use of non-free-flowing UO_2 powders coming from a wet conversion (for example, ex-ADU or ammonium diuranate powder) or from a dry conversion (said conversions being known to those skilled in the art);
- addition of 0.2 to 0.5% of zinc stearate and 0 to 1% of an AZB pore former (known to those skilled in the art);
- pelletizing compression (step 6) at a pressure between 400 and 700 MPa;
- sintering (step 7) for at least 4 hours at a temperature between 1600 and 1760°C, in an

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argon atmosphere containing 5% hydrogen, with an H_2/H_2O ratio of 10 to 30; and - dry centerless grinding (step 8).

By way of nonlimiting example, the compression step (step 30) may be carried out at a pressure of between 50 and 200 MPa, this being tailored according to the characteristics of the non-free-flowing powder. These pressures are therefore higher than the granulation pressures (4 to 10 MPa) generally used in 10 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25 26 27 28 29 30 31 32 33 34 35 36 37 38 39 40 41 42 43 44 45 46 47 48 49 50 51 52 53 54 55 56 57 58 59 60 61 62 63 64 65 66 67 68 69 70 71 72 73 74 75 76 77 78 79 80 81 82 83 84 85 86 87 88 89 90 91 92 93 94 95 96 97 98 99 100 101 102 103 104 105 106 107 108 109 110 111 112 113 114 115 116 117 118 119 120 121 122 123 124 125 126 127 128 129 130 131 132 133 134 135 136 137 138 139 140 141 142 143 144 145 146 147 148 149 150 151 152 153 154 155 156 157 158 159 160 161 162 163 164 165 166 167 168 169 170 171 172 173 174 175 176 177 178 179 180 181 182 183 184 185 186 187 188 189 190 191 192 193 194 195 196 197 198 199 200 201 202 203 204 205 206 207 208 209 210 211 212 213 214 215 216 217 218 219 220 221 222 223 224 225 226 227 228 229 230 231 232 233 234 235 236 237 238 239 240 241 242 243 244 245 246 247 248 249 250 251 252 253 254 255 256 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Also by way of nonlimiting example, the aforementioned tablets may be crushed in one or more jaw crushes or roll mills of 200-250 μm aperture. This crushing may be followed by sieving if the crusher lets through, or runs the risk of letting through, granules having a size greater than 250 μm . The fines possibly resulting from the crushing may usefully be incorporated as raw material into the first blending operation (step 1).

25 By way of yet another nonlimiting example, the
operation of forcing the powder through a sieve (step
29) may be carried out in a machine of the kind used in
MIMAS-type processes (step 3) to fill the first blend
(after the micronization of step 2) before the second
30 blending (step 4). Such machines, which combine
agglomeration/precompaction upstream of the sieve and
control of the maximum granule size by passing the
powder through this same sieve, may produce granules of
the desired characteristics directly.

35 Experience has shown the Applicant that a non-free-flowing powder treated according to the process forming the subject matter of the invention can be used in existing MOX manufacturing plants, by adjusting the parameters of this second blending operation (step 4),

the pelletizing (step 6) and the sintering (step 7), within the adjustment limits routinely used to optimize the manufacturing process according to the characteristics of the various free-flowing UO_2 powders 5 used for MOX fuel manufacture.

The process of the invention therefore makes it possible to extend the range of UO_2 powders which can be used to manufacture MOX fuel, without loosing the benefit of the similarity between the MOX fuel produced 10 according to the invention and the UO_2 fuel manufactured on an industrial scale by the processes known hitherto, starting from the same non-free-flowing UO_2 powder.

It should be understood that the present 15 invention is in no way limited to the methods of implementation described above and that many modifications may be made thereto without departing from the scope of the claims given hereafter.

The non-free-flowing UO_2 conditioning process 20 may especially be applied to UO_2 coming from a conversion other than the conversion of uranium hexafluoride into UO_2 .